P_2O_5/SiO_2 an efficient, green and heterogeneous catalytic system for the solvent-free synthesis of *N*-sulfonyl imines

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Abstract

Silica-supported P_2O_5 (P_2O_5/SiO_2) was used as an efficient, green and cheap catalytic system for the synthesis of *N*-sulfonyl imines via condensation of sulfonamides with several aldehydes under solvent-free conditions. The reactions proceeded rapidly at 110 °C and the desired products were obtained in high to excellent yields.

Keywords: P_2O_5/SiO_2, solvent-free, *N*-sulfonyl imine, sulfonamide, aldehyde

Introduction

Imines bearing electron-withdrawing *N*-substituents are useful intermediates in organic synthesis.[1] Among them, *N*-sulfonyl imines are the centre of attention for organic chemists because the sulfonyl moiety has proven to be a powerful activating group of the C=N bond in these compounds. As a consequence, *N*-sulfonyl imines have been widely used in organic synthesis.[2] In addition, they are excellent substrates in nucleophilic additions,[3] reductions,[4] aza Diels-Alder reactions,[5] aziridine[6] and oxaziridine synthesis[7] as well as ene reactions.[8] Several kinds of synthetic routes toward *N*-sulfonyl imines have been developed namely via the Lewis acid catalyzed reactions of sulfonamides with aldehyde precursors,[9] rearrangement of oxime *O*-sulfinates,[10] tellurium mediated reaction of aldehydes with chloramines T by utilization of *in situ* generated *N*,*N*′-ditosyltellurodiimide,[11] application of *N*-sulfinyl sulfonamides instead of sulfonamides to generate sulfonyl imine *in situ via* a [2+2] cycloaddition and extrusion of sulfur dioxide,[12] generation of sulfonamidosulfones and basic elimination,[13] and catalyzed isomerization or rearrangement of *N*-sulfonyl aziridines.[14]

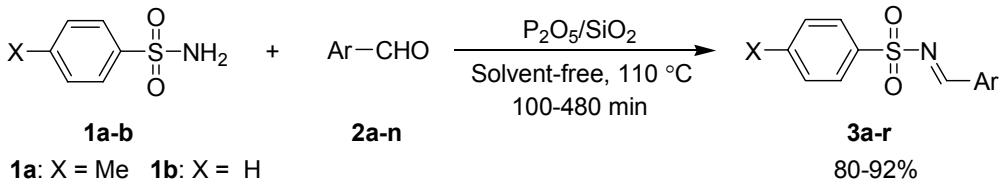
It is worth noting that the methods that have been established for the preparation of *N*-sulfonyl imines are associated with one or more of the following drawbacks: (i) long reaction times, (ii) unsatisfactory yields, (iii) formation of toxic byproducts, and (iv) the use of expensive

and hazardous reagents. Furthermore, some other procedures need cumbersome experimental and multi-step procedure. Therefore, it seems highly desirable to find a one step, and inexpensive protocol for the synthesis of *N*-sulfonyl imines. The aim of this work is to report a simple protocol that makes these valuable building blocks readily available.

As it is known Green Chemistry (a simple definition) is recommended with the corresponding reference. In this sense, heterogeneous organic reactions have many advantages, such as ease of handling, low corrosion, minimum execution time, environmentally safe disposal and waste minimization by developing cleaner synthesis routes.[15] Another important goal in green chemistry is represented by the elimination of volatile organic solvents, in fact, solvent-free conditions that makes synthesis simpler, saves energy, and prevents solvent waste, hazards, and toxicity.[16] Consequently, it is important to note that the combination of heterogeneous catalysis with the use of solventless conditions represent a suitable way toward the so-called ideal synthesis.[17]

Silica-supported phosphorus pentoxide (P_2O_5/SiO_2) is an inexpensive, heterogeneous and commercially available catalytic system which has been used in several transformations, such as condensation of indoles with carbonyl compounds,[18] oxidation of sulfides to the corresponding sulfoxides,[19] nitration of aromatic compounds,[20] conversion of aldehydes to nitriles,[21] acylation of amines,[22] deprotection of 1,1-diacetates,[23] Fries rearrangement,[24] and acetalization of carbonylic compounds.[25]

Considering the above subjects and also along with our previous studies on green organic synthesis,[9a,18,26] herein, is reported an efficient, green and simple method for the production of a set of eighteen *N*-sulfonyl imines from benzene, *p*-toluene sulfonamides and aromatic aldehydes in the presence of P_2O_5/SiO_2 under solvent-free conditions, Scheme 1.



Scheme 1. Condensation of sulfonamides with aldehydes.

Results and Discussion

In order to optimize the reaction conditions, the condensation of 4-methylbenzenesulfonamide with benzaldehyde was studied as a model reaction to provide compound **3a** (Scheme 1). At a first scope, the reaction was examined in the presence of catalytic amount of P_2O_5 at 110 °C under solvent-free conditions; however, the product was obtained with low yield even after prolonging the reaction time (Table 1, entry 1). In these conditions, a sticky reaction mixture was obtained. The yield was remarkably increased when P_2O_5 supported on silica gel (P_2O_5/SiO_2)

was applied as catalyst (Table 1, entry 2). The influence of other supports on the reaction was also investigated (Table 1), as it can be seen, silica gel promoted higher yields. Therefore, silica gel was the support of choice for all reactions. It is also worth mentioning that the model reaction was also carried out in the presence of silica gel without P_2O_5 under solventless conditions at 110 °C, however compound **3a** was afforded in trace yield within 6 hours.

Table 1. The effect of different supports on condensation of 4-methylbenzenesulfonamide with benzaldehyde in the presence of P_2O_5

Entry	Support	Time (min)	Yield[a] (%)
1[b]	-	240	55
2	Silica gel	120	91
3	Neutral alumina	120	72
4	Acidic alumina	120	79
5	Basic alumina	120	65
6	Graphite	120	59

[a]Isolated pure product. [b]Without support.

To compare the efficiency as well as capacity of the solvent-free conditions with respect to solution conditions, the model reaction was examined in the presence of silica-supported P_2O_5 in several solvents; the corresponding results are depicted in Table 2. As it can be seen, the solvent-free method is more efficient.

Table 2. Comparative the reaction of 4-methylbenzenesulfonamide with benzaldehyde in the presence of P_2O_5/SiO_2 using solution conditions versus the solvent-free method

Entry	Solvent	Temperature (°C)	Time (min)	Yield[a] (%)
1[b]	-	110	120	91
2	DMSO	110	300	56
3	DMF	110	300	44
4	CH_3CN	Reflux	720	35
5	CH_2Cl_2	Reflux	720	12
6	$CHCl_3$	Reflux	720	18
7	THF	Reflux	720	26

[a]Isolated pure product. [b]The Solvent-free conditions.

The efficiency and applicability of this new procedure was compared with some reported methods for the preparation of *N*-sulfonyl imines (Table 3). As Table 3 demonstrates, our method afforded the better results.

In order to establish the generality of this method, the benzene and *p*-toluene sulfonamides were condensed with various aldehydes to furnish *N*-sulfonyl imines in high to excellent yields in relatively short reaction times; the respective results are displayed in Table 4.

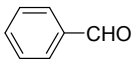
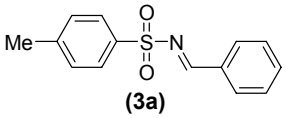
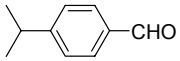
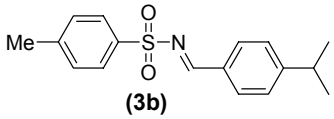
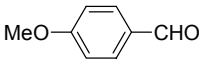
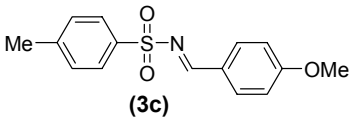
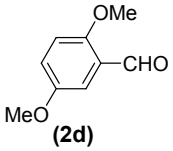
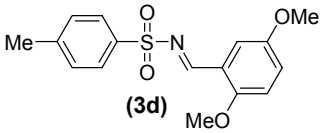
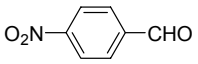
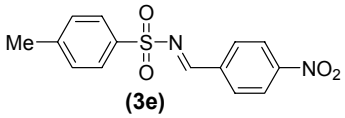
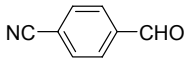
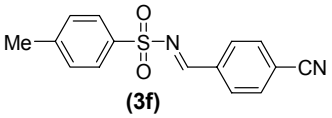
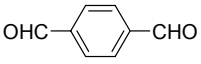
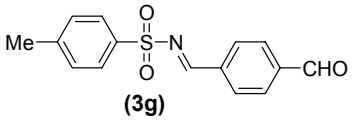
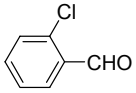
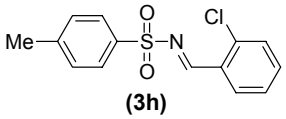
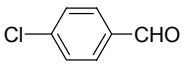
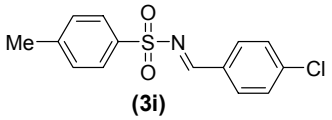
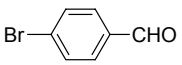
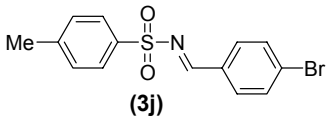
Table 3. The comparative synthesis of compound **3a** using the reported methods versus the present method

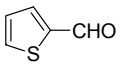
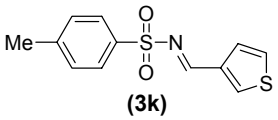
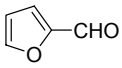
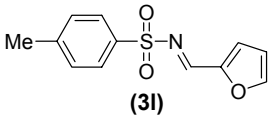
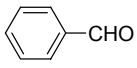
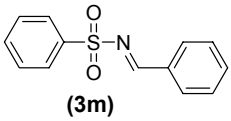
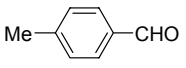
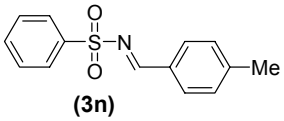
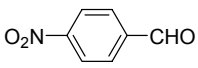
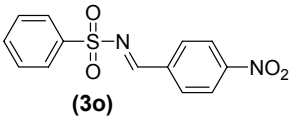
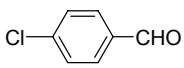
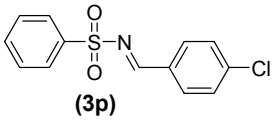
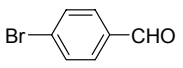
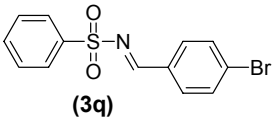
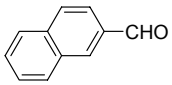
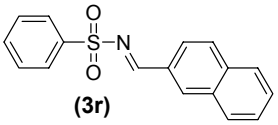
Entry	Reagent and Conditions	Time (min)	Yield[a] (%)	Ref.
1[b]	P_2O_5/SiO_2, solvent-free, 110 ˚C	120	91	-
2	Silica chloride, solvent-free, 120 ˚C	180	75	9a
3	$Si(OEt)_4$/160 ˚C	360	68	9b
4	$CaCO_3$, K10 Clay, $CH(OMe)_3$/Microwave	6	69	9d
5	$TiCl_4$, NEt_3/0 ˚C (CH_2Cl_2)	25	58	9e
6[c]	a) PhCH=NOH b) TsCN, NEt_3/0 ˚C (CCl_4)	30	59	2b
7	PhCHO + Ph_3P=NTs/$RuCl_2(PPh_3)_3$ (CH_2Cl_2)	36	75	27

[a]Isolated pure product.

The influence of both electron-releasing and electron-withdrawing substituents on the aromatic ring of aldehydes upon results of the reaction was investigated. The results showed that electron-releasing substituents had no significant effect on the reaction times and the yields (Table 4, entries 2-4 and 14). However, the presence of electron-withdrawing substituents on the aromatic ring of aryl aldehydes increased the reaction times and decreased the yields (Table 4, entries 5-7 and 15). Moreover, the presence of a halogen on the aromatic ring of aldehydes had negligible effect on the reaction yields; however, these substituents increased the reaction times (Table 4, entries 8-10, 16 and 17).

Table 4. Preparation of *N*-sulfonyl imines from sulfonamides and aldehydes

Entry	Aldehyde	Product	Time (min)	Yield[a] (%)
1	 **(2a)**	 **(3a)**	120	91
2	 **(2b)**	 **(3b)**	120	92
3	 **(2c)**	 **(3c)**	120	90
4	 **(2d)**	 **(3d)**	150	87
5	 **(2e)**	 **(3e)**	480	82
6	 **(2f)**	 **(3f)**	480	84
7[b]	 **(2g)**	 **(3g)**	300	80
8	 **(2h)**	 **(3h)**	240	87
9	 **(2i)**	 **(3i)**	180	89
10	 **(2j)**	 **(3j)**	180	90

11	 (2k)	 (3k)	100	91
12	 (2l)	 (3l)	100	88
13	 (2a)	 (3m)	120	88
14	 (2m)	 (3n)	120	90
15	 (2e)	 (3o)	480	80
16	 (2i)	 (3p)	240	87
17	 (2j)	 (3q)	240	86
18	 (2n)	 (3r)	120	86

[a]Isolated pure product. [b]In this reaction, bis-*N*-sulfonyl imine was obtained in very low yield.

Conclusions

In conclusion, an efficient method for the synthesis of *N*-sulfonyl imines via condensation of sulfonamides with aldehydes, has been developed. This new strategy has several advantages, such as high yields, relatively short reaction times, low cast, simple experimental and as isolation procedures, and finally, it is in agreement with the green chemistry protocols.

Experimental Section

General Procedures. All chemicals were purchased from Merck or Fluka chemical companies. Silica gel 60, 0.063-0.200 mm (70-230 mesh ASTM) was used as support. All known compounds were identified by comparison of their melting points and ^{1}H NMR data with the authentic samples. The ^{1}H NMR (250 MHz) spectra were run on a Bruker Avanced DPX-250, FT-NMR spectrometer. Mass spectra were recorded on a Shimadzu GC MS-QP 1000 EX apparatus. Microanalyses were performed on a Perkin-Elmer 240-B microanalyzer. Melting points were recorded on a Büchi B-545 apparatus in open capillary tubes.

Preparation of P_2O_5/SiO_2 catalytic system. A mixture of SiO_2 (2 g) and P_2O_5 (1 mmol, 0.142 g) was ground vigorously to give P_2O_5/SiO_2 catalytic system as a white powder (2.142 g).

Condensation of sulfonamides with aldehydes under solvent-free conditions. To a well ground mixture of sulfonamide (1 mmol) and P_2O_5/SiO_2 (0.536 g) in a 10 mL round-bottomed flask connected to a reflux condenser was added aldehyde (1.1 mmol). The mixture was stirred in an oil-bath (110 °C) for the times reported in Table 4. Afterward, the reaction mixture was cooled to room temperature and the solid mixture was poured on a Celite pad and washed with acetone (20 mL). The solvent was evaporated and the crude product was dissolved in warm ethyl acetate (2 mL), treated with *n*-hexane (6 mL), and was allowed to stand at room temperature for 5-6 h. During this time, the target molecules were produced and then collected by filtration, washed with *n*-hexane and dried.

(E)-*N*-Benzylidene-4-methylbenzenesulfonamide (3a). Colorless solid; yield: 0.237 g (91%); mp 108-109 °C (Lit.[9a] mp 108 °C); ^{1}H NMR ($CDCl_3$): δ 2.33 (3H, s), 7.25 (2H, d, *J* = 8.2 Hz), 7.48 (2H, t, *J* = 7.8 Hz), 7.51 (1H, t, *J* = 6.2 Hz), 7.80-7.84 (4H, m), 8.99 (1H, s).

(E)-*N*-(4-Isoperopyl-benzylidene)-4-methylbenzenesulfonamide (3b). Colorless solid; yield: 0.278 g (92%); mp 110-112 °C (Lit.[28] mp 113-115 °C); ^{1}H NMR ($CDCl_3$): δ 1.20 (6H, d, *J* = 7 Hz), 2.33 (3H, s), 2.93 (1H, m), 7.18-7.80 (8H, m), 8.91 (1H, s).

(E)-*N*-(4-Methoxy-benzylidene)-4-methylbenzenesulfonamide (3c). Colorless solid; yield: 0.260 g (90%); mp 127-129 °C (Lit.[11] mp 128-129 °C); ^{1}H NMR ($CDCl_3$): δ 2.36 (3H, s), 3.78 (3H, s), 6.93 (2H, d, *J* = 8.5 Hz), 7.29 (2H, d, *J* = 7.8 Hz), 7.81 (2H, d, *J* = 7.5 Hz), 7.85 (2H, d, *J* = 7.6 Hz), 8.90 (1H, s).

(E)-*N*-(2,5-Dimethoxy-benzylidene)-4-methylbenzenesulfonamide (3d). Colorless solid; yield: 0.277 g (87%); mp 125-127 °C (Lit.[9a] mp 124-126 °C); ^{1}H NMR (DMSO-d_6): δ 2.39 (3H, s), 3.69 (3H, s), 3.93 (3H, s), 7.14-7.82 (7H, m), 9.30 (1H, s).

(E)-*N*-(4-Nitro-benzylidene)-4-methylbenzenesulfonamide (3e). Yellow solid; yield: 0.250 g (82%); mp 164-166 °C (Lit.[29] mp 162-164 °C); ^{1}H NMR ($CDCl_3$): δ 2.39 (3H, s), 7.29 (2H, d, *J* = 7.8 Hz), 7.78 (2H, d, *J* = 7.8 Hz), 7.95 (2H, d, *J* = 8.5 Hz), 8.16 (2H, d, *J* = 8.5 Hz), 9.17 (1H, s).

(E)-*N*-(4-Cyanobenzylidene)-4-methylbenzenesulfonamide (3f). Colorless needles; yield: 0.238 g (84%); mp 171-172 °C (Lit.[9a] mp 172-173 °C) (9a); ^{1}H NMR ($CDCl_3$): δ 2.35 (3H, s), 7.28 (2H, d, *J* = 10.2 Hz), 7.66-7.96 (6H, m) 8.97 (1H, s).

(E)-*N*-(4-Formylbenzylidene)-4-methylbenzenesulfonamide (3g). Colorless solid; yield: 0.230 g (80%); mp 98-100 °C; ^{1}H NMR (DMSO-d_6): δ 2.39 (3H, s), 7.25 (2H, d, *J* = 7.6 Hz), 7.33 (2H, d, *J* = 8.0 Hz), 7.72 (2H, d, *J* = 8.0 Hz), 8.08 (2H, d, *J* = 7.6 Hz), 9.22 (1H, s), 10.12 (1H, s); MS *m/z*: 287 (M^+); Anal. calcd. for $C_{15}H_{13}NO_3S$: C, 62.70; H, 4.56; N, 4.87. Found: C, 62.89; H, 4.40; N, 5.07.

(E)-*N*-(2-Chloro-benzylidene)-4-methylbenzenesulfonamide (3h). Colorless needles; yield: 0.256 g (87%); mp 127-128 °C (Lit.[9b] mp 128-129 °C); ^{1}H NMR ($CDCl_3$): δ 2.43 (3H, s), 7.29-7.90 (8H, m), 9.13 (1H, s).

(E)-*N*-(4-Chlorobenzylidene)-4-methylbenzenesulfonamide (3i). Colorless needles; yield: 0.262 g (89%); mp 169-170 °C (Lit.[11] mp 172-173 °C); ^{1}H NMR ($CDCl_3$): δ 2.46 (3H, s), 7.15 (2H, d, *J* = 9.1 Hz), 7.38 (2H, d, *J* = 9.0 Hz), 7.86 (4H, m), 9.01 (1H, s).

(E)-*N*-(4-Bromobenzylidene)-4-methylbenzenesulfonamide (3j). Colorless solid; yield: 0.305 g (90%); mp 169-170 °C (Lit.[2a] mp 172-173 °C); ^{1}H NMR ($CDCl_3$): δ 2.50 (3H, s), 7.26-7.85 (8H, m), 9.04 (1H, s).

(E)-4-Methyl-*N*-(thiophen-3-ylmethylene)benzenesulfonamide (3k). Colorless needles; yield: 0.241 g (91%); mp 128-130 °C (Lit.[9a] mp 127-129 °C); ^{1}H NMR ($CDCl_3$): δ 2.43 (3H, s), 7.29-8.15 (7H, m), 9.01 (1H, s).

(E)-*N*-(Furan-2-ylmethylene)-4-methylbenzenesulfonamide (3l). Brown solid; yield: 0.220 g (88%); mp 99-100 °C (Lit.[11] mp 101 °C); ^{1}H NMR ($CDCl_3$): δ 2.32 (1H, s), 6.76-7.75 (7H, m), 8.83 (1H, s).

(E)-*N*-Benzylidenebenzenesulfonamide (3m). Colorless solid; yield: 0.215 g (88%); mp 76-78 °C (Lit.[9a] mp 77-80 °C); ^{1}H NMR ($CDCl_3$): δ 7.61 (6H, m), 8.02 (4H, m), 9.05 (1H, s).

(E)-*N*-(4-Methylbenzylidene)benzenesulfonamide (3n). Colorless solid; yield: 0.234 g (90%); mp 113-115 °C (Lit.[9c] mp 116-118 °C); ^{1}H NMR ($CDCl_3$): δ 2.38 (3H, s), 7.21 (2H, d, *J* = 7.3 Hz), 7.46 (2H, m), 7.51 (1H, m), 7.76 (2H, d, *J* = 7.3 Hz), 7.93 (2H, m), 9.18 (1H, s).

(E)-*N*-(4-Nitrobenzylidene)benzenesulfonamide (3o). Yellow solid; yield: 0.233 g (80%); mp 164-165 °C (Lit.[29] mp 162-164 °C); ^{1}H NMR ($CDCl_3$): δ 7.41 (2H, m), 7.48 (1H, m), 7.91 (4H, m), 8.19 (2H, d, *J* = 8.7 Hz), 9.15 (1H, s).

(E)-*N*-(4-Chlorobenzylidene)benzenesulfonamide (3p). Colorless solid; yield: 0.243 g (87%); mp 131-133 °C (Lit.[9c] mp 127-130 °C); ^{1}H NMR ($CDCl_3$): δ 7.36 (2H, d, *J* = 7.4 Hz), 7.46 (2H, m), 7.53 (1H, m), 7.83 (2H, d, *J* = 7.4 Hz), 7.96 (2H, m), 9.10 (1H, s).

(E)-*N*-(4-Bromobenzylidene)benzenesulfonamide (3q). Colorless solid; yield: 0.278 g (86%); mp 205-207 °C (Lit.[28] mp 208-209 °C); ^{1}H NMR ($CDCl_3$): δ 7.37 (2H, d, *J* = 8.0 Hz), 7.44 (2H, m), 7.55 (1H, m), 7.78 (2H, d, *J* =8.0 Hz), 7.97 (2H, m), 9.14 (1H, s).

(E)-*N*-(Naphthalen-2-ylmethylene)benzenesulfonamide (3r). Pale yellow solid; yield: 0.255 g (86%); mp 166-168 °C (Lit.[30] 172-174 °C); ^{1}H NMR ($CDCl_3$): δ 7.46-7.60 (5H, m), 7.79-7.94 (4H, m), 7.98 (2H, d, *J* = 8.0 Hz), 8.27 (1H, s), 9.13 (1H, s).

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